

REPORT DOCUMENTATION PAGE

Form Approved OMB NO. 0704-0188

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1. AGENCY USE ONLY (Leave Blank)	2. REPORT DATE:	3. REPORT TYPE AND DATES COVERED Final Report 15-Aug-2004 - 31-Jan-2006	
4. TITLE AND SUBTITLE Ab initio relativistic study of formation of ultracold polar molecules		5. FUNDING NUMBERS W911NF0410397	
6. AUTHORS Svetlana Kotochigova	8. PERFORMING ORGANIZATION REPORT NUMBER		
7. PERFORMING ORGANIZATION NAMES AND ADDRESSES Temple University Sponsored Projects Administration 1601 N Broad Street Philadelphia, PA 19122 -			
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) U.S. Army Research Office P.O. Box 12211 Research Triangle Park, NC 27709-2211	10. SPONSORING / MONITORING AGENCY REPORT NUMBER 47083-PH.1		
11. SUPPLEMENTARY NOTES The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.			
12. DISTRIBUTION AVAILABILITY STATEMENT Approved for Public Release; Distribution Unlimited	12b. DISTRIBUTION CODE		
13. ABSTRACT (Maximum 200 words) The abstract is below since many authors do not follow the 200 word limit			
14. SUBJECT TERMS Alkali-metal molecules, ultra-cold polar molecules, dynamic polarizability, decoherence		15. NUMBER OF PAGES Unknown due to possible attachments	
		16. PRICE CODE	
17. SECURITY CLASSIFICATION OF REPORT UNCLASSIFIED	18. SECURITY CLASSIFICATION ON THIS PAGE UNCLASSIFIED	19. SECURITY CLASSIFICATION OF ABSTRACT UNCLASSIFIED	20. LIMITATION OF ABSTRACT UL

Report Title

Ab initio relativistic study of formation of ultracold polar molecules.

ABSTRACT

We determine conditions for effective quantum control over internal and external degrees of freedom of polar alkali-metal molecules in an optical trap. We calculate the dynamic molecular polarizability of KRb and RbCs as a function of laser frequency. The real part of the molecular polarizability determines the depth of the trapping potential while the imaginary part contributes to laser-induced decoherences. Our results show that a relatively modest laser intensity is needed for strong confinement of polar molecules in an optical trap and that decoherence effects are manageable.

More complete information on the scientific progress for the period of the extension is given in the Attachment.

List of papers submitted or published that acknowledge ARO support during this reporting period. List the papers, including journal references, in the following categories:

(a) Papers published in peer-reviewed journals (N/A for none)

1. S. A. Kotochigova, K. P. Kirby, N. S. Brickhouse, P. J. Mohr, and I. I. Tupitsyn, "X-ray Diagnostics of astrophysical plasmas" AIP, pp.161-171 (2005).
2. U. D. Jentschura, S. Kotochigova, E.-O. Le Bigot, P. J. Mohr, and B.N. Taylor, Phys. Rev. Lett. 95, 163003 (2005).
3. S. Kotochigova and E. Tiesinga, J. Chem. Phys. 123, 174304 (2005).
4. E. Ahmed, A. Hansson, P. Qi, L. Li, T. Kirova, J. Qi, A. Lazoudis, S. Magnier, S. Kotochigova, and M. Lyyra, J. Chem. Phys. accepted for publication in (2005).

Number of Papers published in peer-reviewed journals: 4.00

(b) Papers published in non-peer-reviewed journals or in conference proceedings (N/A for none)

1. S. Kotochigova and E. Tiesinga, "Controlling ultracold polar molecules in optical lattices. arXiv: quant-physics/0602017, (2006).

Number of Papers published in non peer-reviewed journals: 1.00

(c) Papers presented at meetings, but not published in conference proceedings (N/A for none)

1. S. Kotochigova, "Ab initio study of polar molecules in optical lattices" Joint Workshop on Theory of Molecules, Telluride, CO (2005).
2. S. Kotochigova, "Controlling polar molecules in optical lattices", Workshop: Quantum Computing with Polar Molecules Arlington, VA (2005).
3. S. Kotochigova, "Creating polar molecules in optical lattices", Yale University, Physics Department, December (2005).

Number of Papers not Published: 3.00

(d) Manuscripts

1. S. Kotochigova and E. Tiesinga, "Controlling ultracold polar molecules in optical lattices", submitted to PRL (2005).
2. E. Ahmed, A. Hansson, P. Qi, L. Li, T. Kirova, J. Qi, A. Lazoudis, S. Magnier, S. Kotochigova, and M. Lyyra, J. Chem. Phys. accepted for publication in (2005).

Number of Manuscripts: 2.00

Number of Inventions:

Graduate Students

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
FTE Equivalent:	
Total Number:	

Names of Post Doctorates

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>	National Academy Member
Dr. Svetlana Kotochigova	1.00	No
FTE Equivalent:	1.00	
Total Number:	1	

Names of Under Graduate students supported

<u>NAME</u>	<u>PERCENT SUPPORTED</u>
FTE Equivalent:	
Total Number:	

Names of Personnel receiving masters degrees

<u>NAME</u>
Total Number:

Names of personnel receiving PhDs

<u>NAME</u>
Total Number:

Names of other research staff

NAME

PERCENT SUPPORTED

FTE Equivalent:

Total Number:

Sub Contractors (DD882)

Inventions (DD882)

Ab Initio Relativistic Study of Formation of Ultracold Polar Molecules

Svetlana Kotochigova

I. STATEMENT OF THE PROBLEM STUDIED

In this report we provide theoretical information on polar KRb and RbCs molecules, which are proposed as good candidates for ultracold experiments. Our goal is to realistically estimate the effect of optical dipole traps and photoassociation laser fields on ultra-cold molecules formed by Raman photoassociation. We assume production of translationally cold molecules by a stimulated Raman process to specific target ro-vibrational levels of electronic excited states. In two-color Raman photoassociation the effect of excited state spontaneous decay can be strongly suppressed. Finally, we perform a quantitative estimate of the dynamic polarizability of alkali-metal polar molecules in an optical domain to determine a depth of the trapping potential as well as laser induced decoherence.

II. SUMMARY OF THE MOST IMPORTANT RESULTS

We start with a molecule in the lowest rovibrational level of the ground electronic potential, created by Raman photoassociation from its individual atoms. We envision that the photoassociation experiment might be done in an optical dipole trap where a focussed laser beam holds both atoms and molecules.

An important aspect of photoassociative formation of ultracold molecules is the behavior of molecules in presence of optical dipole traps and photoassociation laser fields. If an optical dipole or photoassociation laser (with a frequency between 800 nm to 1000 nm) is nearly resonant to some molecular transition, this leads to transfer of population from the lowest rovibrational level of the ground potential to the rovibrational level of an excited potential, which then by spontaneous emission can decay to many rovibrational levels of the ground potential. As a result, we lose control over the molecule in a trap. In addition, spontaneous decay can cause a momentum transfer and change the position of the center of mass of the molecule, leading to a motional decoherence. This effect might be small as the distance traveled by the molecule over the natural life-time of the rovibrational state of an excited

potential is small. However, this needs to be confirmed. On the other hand, when a molecule is in the electronic excited state and has a different polarizability than in the ground state, which in turn implies a different trapping potential, we can again loose the molecule from the trap.

The property of a molecule relevant to photoassociation, that can help us to understand coherent and decoherent effects in an optical trap, is the molecular dynamic polarizability as a function of laser frequency. The real part of the molecular polarizability determines the depth of the trapping potential while the imaginary part contributes to laser-induced decoherences. The contribution to the dynamic polarizability of the ground state is due to the dipole coupling to other rovibronic states of the ground and excited potentials.

Assuming that the alkali-metal molecule is in a rovibrational state of the ground $X^1\Sigma^+$ potential, its dynamic polarizability in SI units is given in terms of the dipole coupling to other rovibrational states of the ground and excited potentials as

$$\alpha(h\nu, \vec{\epsilon}) = \frac{1}{\epsilon_0 c} \sum_f \frac{(E_f - ih\gamma_f/2 - E_i)}{(E_f - ih\gamma_f/2 - E_i)^2 - (h\nu)^2} \times |\langle f | d \hat{R} \cdot \vec{\epsilon} | i \rangle|^2. \quad (1)$$

where c is the speed of light, ϵ_0 is the electric constant, \hat{R} is the orientation of the interatomic axis, i and f denote the initial $|vJM\rangle$ and intermediate $|v'J'M'\rangle$ rovibrational wavefunctions of the $|X^1\Sigma^+\rangle$ and $|\Omega\rangle$ electronic states, respectively. Here, Ω labels either the $X^1\Sigma^+$ state or any excited state and $\langle f | d | i \rangle$ are R -dependent permanent or transition electronic dipole moments. The quantities M and M' are the projections along a laboratory fixed coordinate system of \vec{J} and \vec{J}' , respectively. The energy E_i is a rovibrational energy in the $X^1\Sigma^+$ state and E_f is the rovibrational energy of the intermediate Ω states. Finally, the line widths γ_f describe the spontaneous and any other decay mechanism that leads to loss of molecules. Equation 1 includes a sum over dipole transitions to higher excited rovibrational levels within the $X^1\Sigma^+$ potential as well as to rovibrational levels of excited potentials. The sum excludes the initial state. Contributions from scattering states or the continuum of the Ω states must also be included. This sum, however, can be truncated assuming that transitions may have zero or near zero electronic dipole moments and/or are far detuned. Nevertheless, a significant number of excited potentials and vibrational states have to be included.

The calculation of the molecular dynamic polarizability includes a summation over dipole transitions to higher excited rovibrational levels within the ground potential as well as

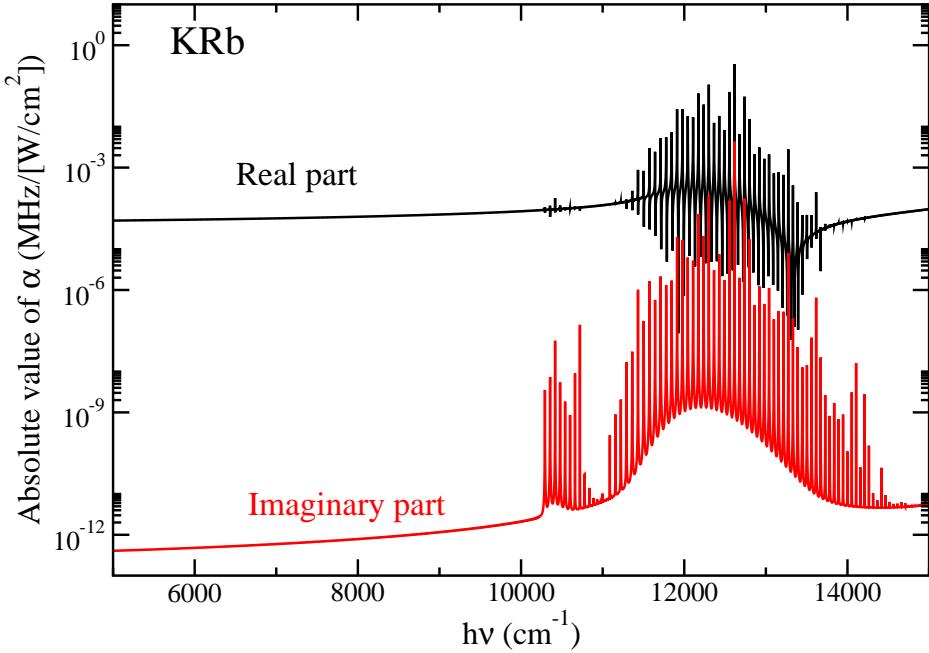


FIG. 1: Real (black line) and imaginary part (red line) of the dynamic polarizability of the $v = 0, J = 0$ level of the $X^1\Sigma^+$ state of KRb as a function laser frequency.

to rovibrational levels of excited potentials and its continua. The precise calculation of molecular polarizability requires knowledge of molecular potential surfaces, transition dipole moments and Franck-Condon factors. We performed ab initio numerical calculations of KRb and RbCs polar molecules using our relativistic configuration-interaction valence bond method for electronic properties combined with a nuclear dynamics method (discrete variable representation DVR for the Schrodinger equation) for the rovibrational motion of the molecules. The relativistic Hamiltonian with Dirac-Fock and Sturmian basis sets allows us to include major relativistic effects nonperturbatively.

Figures ?? show the real and imaginary parts of the polarizability of KRb and RbCs in the infrared and optical domain. The imaginary part of the polarizability is basically due to spontaneous emission from excited states and is much smaller than the real part of polarizability. The ratio is about seven orders of magnitude and decoherence is expected to be small and manageable. The resonant-like features in Figs. ?? are due to bound states of

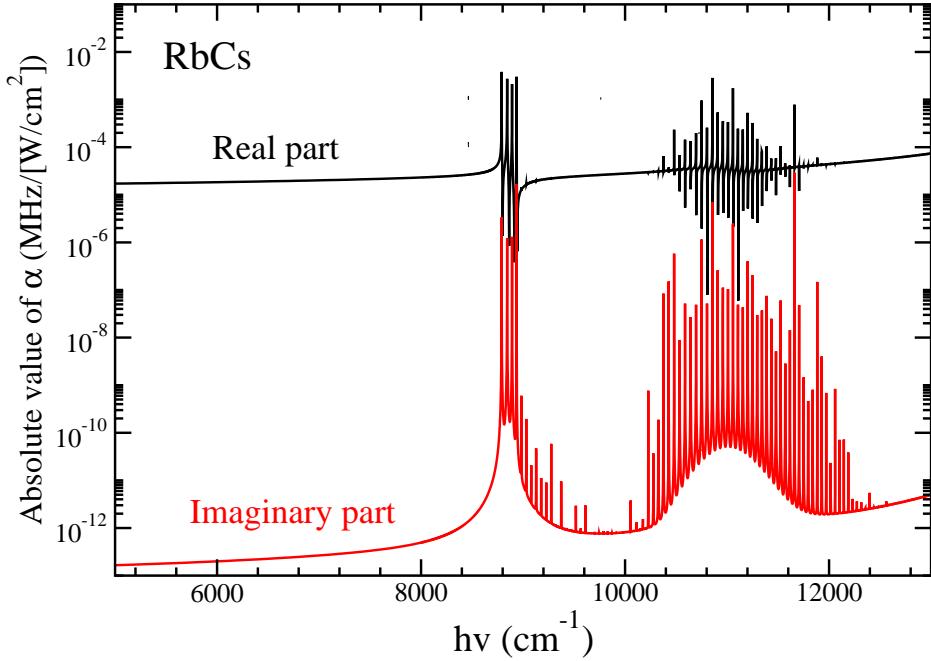


FIG. 2: Real (black line) and imaginary part (red line) of the dynamic polarizability of the $v = 0, J = 0$ level of the $X^1\Sigma^+$ state of RbCs as a function laser frequency. Practical laser frequencies are indicated.

excited potentials and their locations are closely related to the shape of the excited potentials. The vibrational wavefunction of the $v = 0$ and $J = 0$ level of the X state is highly localized around the minimum of the ground state potential and, hence, a large vibrationally averaged transition dipole moment occurs when both the electronic dipole moment is large and excited vibrational levels have good overlap with this level. Rapidly oscillating polarizability above 10000 cm^{-1} for KRb and 9000 cm^{-1} for RbCs are due to dipole transitions to rovibrational levels of the $3(0^+)$ potential. The polarizability below these regions varies relatively slowly and Franck-Condon factors are small.

Using our results we propose two frequency intervals in which resonant excitation is unlikely and are most easy to work with experimentally. We suppose that experimentalists want to stay away from resonant frequencies, as spontaneous emission is large there and we might have unwanted population in electronically-excited vibrational states. We suggest

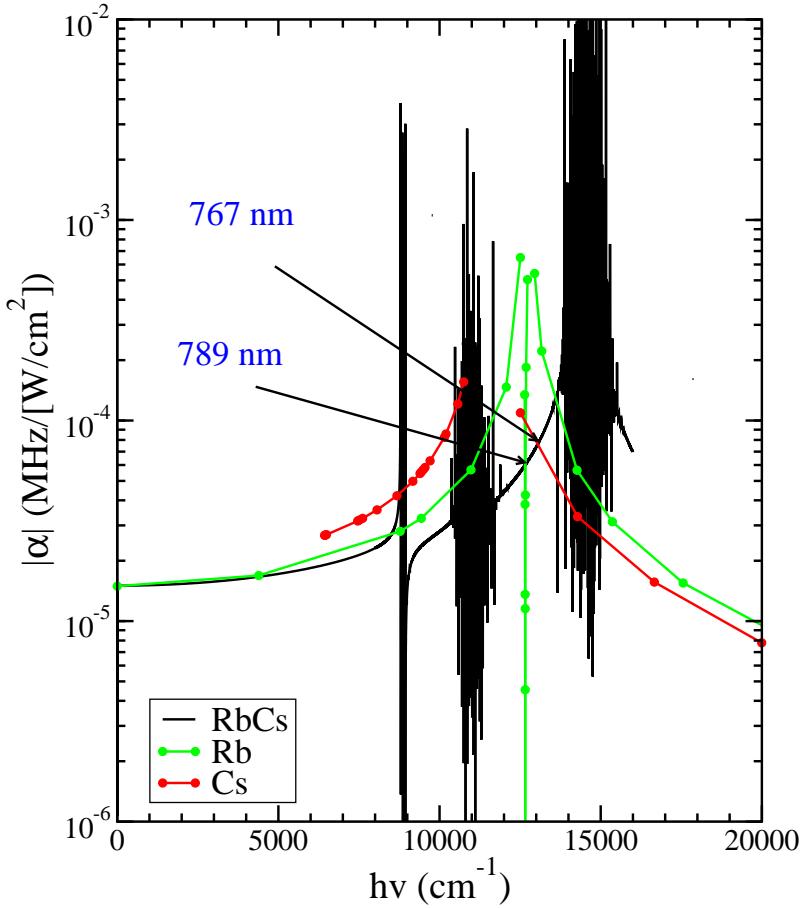


FIG. 3: Comparison of molecular and atomic dynamic polarizabilities of the ground states of RbCs (black curve), Rb (green curve) and Cs (red curve) as a function of laser frequency. Practical laser frequencies are indicated.

lasers with wavelengths between 680 ± 35 nm for a KRb and 790 ± 40 nm for a RbCs experiment. In addition, telecommunication wavelengths between 1.03 ± 0.05 μm seem practical for both molecules.

In addition, it is important to investigate and compare the strength of dipole forces due to the optical trap as seen by free atoms and molecules. For example, we want to find laser frequencies at which a laser field creates the same trapping potential for both colliding atoms and molecule in order to avoid possible loss of molecules during photoassociation. The result of our investigation is shown in Fig. 3. We compared our data for the molecular

polarizability with the published data on the dynamic polarizability of Rb and Cs atoms by Sofronova *et al.* [arXiv:physics/0508087 (2005)]. This comparison allows us to determine so called "magic" wavelengths (between 789 nm and 767 nm), where atoms and molecules have almost the same trapping potentials.